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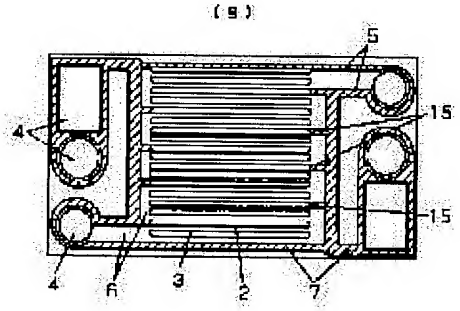
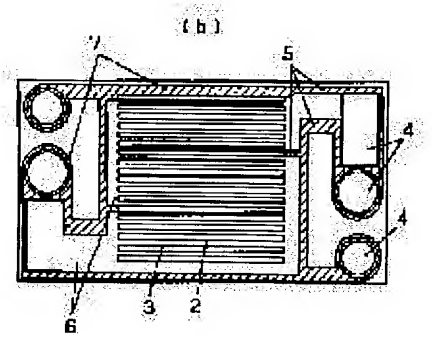
(54) SOLID HIGH POLYMER ELECTROLYTE FUEL CELL

(57)Abstract:

PROBLEM TO BE SOLVED: To reduce cost at mass producing, and to reduce size by providing a pair of electrodes for sandwiching a solid high polymer electrolyte film and a supply discharge means of fuel gas, constituting a conductive separator between laminated cells out of metal having an inactive metallic layer on the surface in an acidic atmosphere, and connecting the gas flowing groove to the gas supply discharge means by a gas sealing material.



SOLUTION: A conductive separator is preferably consists an SUS 316 plate having the surface plate with gold and silver, and on the hydrogen side, a gas guiding phenol groove 6 up to a wavy gas flowing groove is formed of the phenol projecting part 5 from a manifold hole 4 through press working. A curved continuing gas flowing groove is arranged in an air side separator. An electrode/electrolyte jointing body having a positive/negative electrode catalyst layer jointed to both surfaces of the central part of a proton conductive high polymer electrolyte fil is sandwiched by two kinds of separators and a gasket to become a constituting unit of a fuel cell. The separator can be formed thinner than a carbon plate by obviating cutting work.

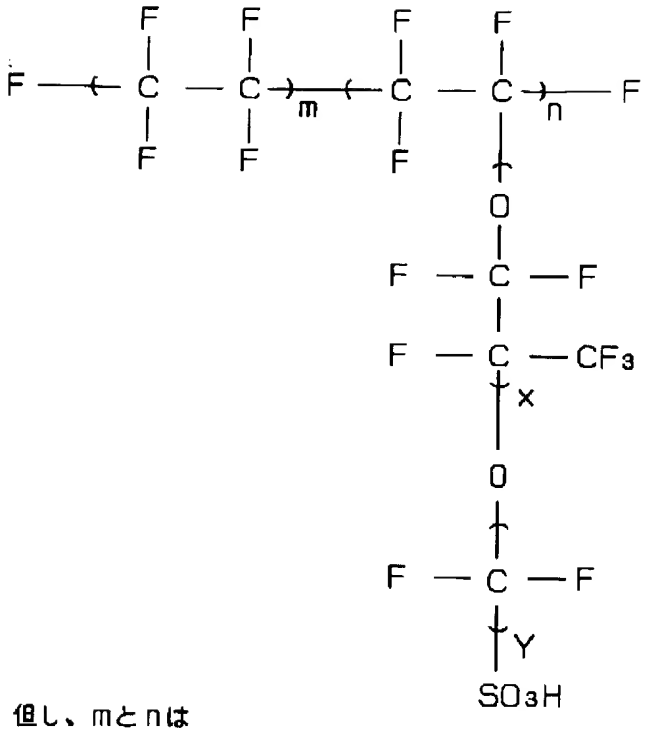


LEGAL STATUS

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] this invention -- the power source for a portable power source and electric vehicles, and domestic -- cogeneration -- it is related with the fuel cell using the solid-state polyelectrolyte used for a system etc.
[0002]
[Description of the Prior Art] The fuel cell using a solid-state polyelectrolyte is making the fuel gas containing hydrogen, and the fuel gas containing oxygen, such as air, react electrochemically, and makes coincidence generate power and heat. The structure forms first the catalytic-reaction layer which uses as a principal component the carbon powder which supported the metal catalyst of a platinum system in both sides of the polyelectrolyte film to which a hydrogen ion is conveyed alternatively. Next, the diffusion layer having the permeability of fuel gas and electronic conductivity is formed in the external surface of this catalytic-reaction layer, this diffusion layer and a catalytic-reaction layer are doubled, and it considers as an electrode.
[0003] Next, the fuel gas to supply leaks outside, or around an electrode, gas-seal material and a gasket are arranged both sides of the polyelectrolyte film so that two kinds of fuel gas may not be mixed mutually. It unites with an electrode and the polyelectrolyte film, and this sealant and gasket are assembled beforehand, and call this MEA (electrode electrolyte membrane zygote). In the outside of MEA, while fixing this mechanically, the conductive separator plate for connecting adjoining MEA to a serial electrically mutually is arranged. Reactant gas is supplied to an electrode surface and the gas passageway for carrying away generation gas and surplus gas is formed in the part in contact with MEA of a separator plate. Although a gas passageway can also be prepared apart from a separator plate the method which prepares a slot on the surface of a separator, and is made into a gas passageway is common.
[0004] In order to supply fuel gas to this slot, it branches in the number of sheets of the separator which uses piping which supplies fuel gas, and the piping fixture which connects that branching place with a direct separator-like slot i needed. An external manifold is called for the type which connects this fixture directly from the charging line of a manifold, and the call and the above fuel gas. There is a thing of the format called the internal manifold which simplified structure more in this manifold. An internal manifold prepares the penetrated hole in the separator plate in which the gas passageway was formed, and supplies direct fuel gas for the entrance of a gas passageway to it from through and this hole even to this hole.
[0005] Since a fuel cell generates heat during operation, in order to maintain a cell in the good temperature condition is necessary to cool by cooling water etc. Usually, although the cooling section which pours cooling water every one 3 cel is inserted between separators, cooling water passage is established in the tooth back of a separator, and it considers as the cooling section in many cases. After it piles up these MEA(s) and separators, and the cooling section by turns and they carry out a 10-200 cel laminating, it is the structure of a common layer built cell which this is inser with an end plate and fixed from both ends with a conclusion bolt through a collecting electrode plate and an electric insulating plate.
[0006] In the fuel cell of such a solid-state macromolecule mold, conductivity of a separator is high, and its gas airtightness is high to fuel gas, and it has the need with high corrosion resistance to the reaction at the time of carryin out oxidation reduction of hydrogen/the oxygen further. Carbon ingredients, such as glassy carbon and expanded graphite, usually constituted the conventional separator from such a reason, and, cutting in the front face, and in the case of expanded graphite, the gas passageway was also produced by molding with a mold.
[0007] However, it replaces with the carbon ingredient used from recent years and the former, and the attempt using metals, such as stainless steel, is performed.
[0008]
[Problem(s) to be Solved by the Invention] By the approach by cutting of the conventional carbon plate, it is difficul with the ingredient cost of a carbon plate to reduce the cost for cutting this, and the approach using expanded graphit also has high ingredient cost, and is considered to be a failure for this utilization.

[0009] Moreover, by the approach using an above-mentioned metal, since a metal is put to the ambient atmosphere o an oxidizing quality at an elevated temperature, if it is used for a long period of time, corrosion and the dissolution o metal will take place. If a metal corrodes, the electric resistance of a corrosion part will increase and the output of a c will decline. Moreover, if a metal dissolves, the dissolved metal ion will be spread in a polyelectrolyte, the trap of th will be carried out to the ion-exchange site of a polyelectrolyte, and own ionic conduction nature of a polyelectrolyte will fall as a result. When the metal was used for the separator as it was and the cell was operated according to these causes for a long period of time, the technical problem that generating efficiency fell gradually occurred.
[0010]
[Means for Solving the Problem] In order to solve the above technical problem the solid-state polyelectrolyte mold f cell of this invention In the solid-state polyelectrolyte mold fuel cell which carried out the laminating of the cell possessing the means which carries out supply discharge of the fuel gas to the electrode of the pair which sandwiche the solid-state polyelectrolyte film, and said electrode through the conductive separator Said conductive separator becomes with the metallic material in which the inactive metal layer to an acid ambient atmosphere was formed on t front face. And it is characterized by said conductive separator connecting the means which carries out supply discharge of said gas circulation slot and said fuel gas with the ingredient which forms the gas circulation slot which circulates said fuel gas, and has the gas-seal nature to said fuel gas further.
[0011] As for the inactive metal layer metal layer to the acid ambient atmosphere formed in the front face of a conductive separator, at this time, consisting of gold or silver is effective.
[0012] Moreover, as for the inactive metal layer metal layer to the acid ambient atmosphere formed in the front face a conductive separator, it is effective to have physical flexibility.
[0013] Moreover, it is effective that the gas circulation slots formed in the conductive separator are two or more parallel straight-line configurations mutually.
[0014] Moreover, it is effective that the heights of the gas circulation slot formed in one field of a conductive separat form the concave of a gas circulation slot at the tooth back of said conductive separator.
[0015] The above manufacture approach is characterized by having washing down stream processing, an activation process, nickel strike plating down stream processing, and gold plate or silver plating down stream processing.
[0016]
[Embodiment of the Invention] The point of this invention is holding down corrosion and the dissolution of a conductive separator made from a metal, and even if it is put to an acid ambient atmosphere, with conductivity havin it is having found out the approach of maintaining chemical inertness.
[0017] Moreover, when using a conductive separator made from a metal for a fuel cell, it is useful that the heights of the gas circulation slot formed in one field of a conductive separator form the concave of a gas circulation slot at the tooth back of a conductive separator. However, when the layer built cell was made using this, the electric contact par of a conductive separator became point contact, and when this was put to the acid ambient atmosphere in the long ru the contact resistance of this part increase-ized. Then, in order to conquer this phenomenon, increase-ization of the above-mentioned contact resistance was conquered by forming the conductive layer which has the physical flexibilit of the shape for example, of sponge into the surface part of a conductive separator.
[0018] Hereafter, the gestalt of operation of this invention is explained, referring to a drawing.
[0019]
[Example] (Example 1) What supported the platinum particle of mean particle diameter 30 [about] 25% of the weig to acetylene black system carbon powder was made into the catalyst of a reaction electrode. The dispersion solution which distributed the powder of the perfluorocarbon sulfonic acid which showed this catalyst powder to the solution which isopropanol was made to distribute by (** 1) in ethyl alcohol was mixed, and it was made the shape of a paste This paste was used as the raw material, it was with screen printing, and the electrode catalyst bed was formed in one field of a carbon nonwoven fabric with a thickness of 250 micrometers. The amount of platinum contained in the reaction electrode after formation adjusted the amount of 0.5 mg/cm2 and perfluorocarbon sulfonic acid so that 1.2mg /might be set to 2 cm.
[0020]
[Formula 1]

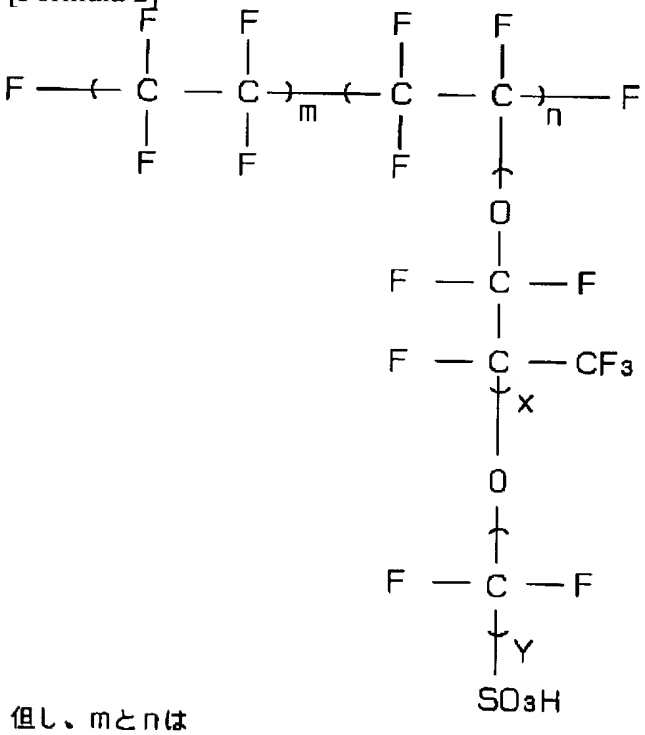


但し、mとnは
m＝5～13．5あたり　n＝1とした。
x＝1
Y＝2

[0021] These electrodes considered the positive electrode and the negative electrode as the same configuration, to bo sides of the core of the proton conductivity polyelectrolyte film which has a somewhat larger area than an electrode, joined with the hotpress so that the printed catalyst bed might touch an electrolyte membrane side, and they created t electrode / electrolyte zygote (MEA) in them. Here, what thin-film-ized the perfluorocarbon sulfonic acid shown in (2) in thickness of 25 micrometers as a proton conductivity polyelectrolyte was used.

[0022]

[Formula 2]



但し、mとnは
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x＝2
Y＝2

0023 he structure f e ch c mp nent f the p lymer electr lyte fuel cell pr duced by this ex mple w s sh wn in

drawing 1 , drawing 2 , and drawing 3 .
[0024] First, the creation approach of the conductive separator which consists of a metal which gold-plated the front face is shown. As shown in drawing 1 , the wavelike processing section 1 of 5.6mm pitch (flute width of about 2.8m was formed in the center-section 10cmx9cm field by press working of sheet metal using SUS316 plate with a thickn of 0.3mm. At this time, the depth (height of a crest 3) of a slot 2 was set to about 1mm. Next, the gold plate layer wa formed in this front face at the thickness of 0.5 micrometers. The plating approach performed activation with the hea sulfuric-acid liquid heated at 80 degrees C after washing to the cleaning agent using the DIPU SOL (trade name of a DIPU SOL company), and performed nickel strike plating processing, nickel-plating processing, and gold plate processing after that. Next, as shown in drawing 1 , the manifold hole 4 for supplying and discharging hydrogen gas, cooling water, and air, respectively was formed in two sides which oppose.
[0025] Next, as shown in drawing 2 (a), the slot 6 which guides gas by the heights 5 made with phenol resin from the manifold hole to the gas circulation slot by processing of a metal plate was established in the separator which becom a hydrogen side. Moreover, the heights 5 made with phenol resin so that two slots might adjoin each other mutually might be curved and connected were piled up.
[0026] The heights made of this phenol resin presupposed that thickness is the same as the height of the crest of the s of a separator plate at about 1mm. It forms also like the periphery section of a separator plate, and the perimeter of a manifold hole, and the gasket 7 corresponding to the configuration of a metal plate is constituted.
[0027] Furthermore, as shown in drawing 2 (b), the separator which becomes an air side formed the gas circulation s where six adjacent slots curved and continued. Structure is changed by the air and hydrogen gas side because quantit of gas flow differ about 25 times by the air and hydrogen gas side. Conversely, if it says, it is possible by changing th configuration of the gas circulation slot made of resin with such structure according to a quantity of gas flow to mak gas pressure loss with the optimal gas flow rate.
[0028] Next, as shown in drawing 3 , on both sides of MEA8, it considered as the configuration unit of a cell with th two kinds of separators, and a gasket. The location of the gas circulation slot 9 by the side of hydrogen and the gas circulation slot 10 by the side of air is constituted so that it may correspond, and it was made not to require the superfluous chinaberry force for an electrode, as drawing 3 showed. The cooling section 11 which pours cooling wat for a cell for every 2 cel laminating was formed. Conductivity and the distributivity of cooling water were secured to the cooling section using the metal mesh 12 made from SUS316, and it considered as the seal section by forming the gasket 7 made of phenol resin in the periphery section and the gas manifold section. The part which needs gas seals, such as a gasket, MEA and a separator plate, a separator plate, and a gasket, a separator plate, secured seal nature by applying grease 13 thinly, without reducing conductivity not much.
[0029] MEA shown above was concluded by the pressure of 20 kgf/cm2 with the veneer and conclusion rod made fr stainless steel through the collecting electrode plate and the electric insulating plate, after carrying out the laminating the 50 cels. Gas leaked, when the bonding pressure force was too small, since contact resistance was also large, the c engine performance became low, but since the electrode was damaged or the separator plate deformed when too conversely large, it was important to change a bonding pressure according to the design of a gas circulation slot.
[0030] What constituted the conductive separator with SUS316 plate which does not carry out a surface coat like the cell of the above-mentioned example as a cell of the example of a comparison was produced. By the cell of the exam of a comparison, it was presupposed except [all] the conductive separator that it is the same as that of the configuration of the above-mentioned example.
[0031] Thus, the produced polyelectrolyte mold fuel cell of this example and the example of a comparison was held 85 degrees C, and the air which humidified and warmed the hydrogen gas humidified and warmed so that a 83-degre C dew-point might come at one electrode side so that a 78-degree C dew-point might come at another electrode side was supplied. Consequently, at the time of no-load [which does not output a current outside], the cell open circuit voltage of 50V was obtained.
[0032] The continuation generation-of-electrical-energy trial was performed for this cell on condition that 80% of fu utilization rates, 40% of ratios of oxygen utilization, and current density 0.5 A/cm2, and time amount change of outp characteristics was shown in drawing 5 . Consequently, compared with an output declining with drive time amount, a for the cell of the example of a comparison, the cell of this example checked maintaining the cell output of 1000W (22V-45A) over 8000 hours or more.
[0033] Although the case where gas circulation slots were two or more parallel straight lines was tried in this examp various structures, such as structure which connects a gas discharge manifold hole from a gas supply manifold throu the bend 14 of 2 times in a gas circulation slot, and structure which connects the manifold hole of a center section an an outside manifold hole like the husks of a snail in a gas circulation slot, are also possible like drawing 4 .
[0034] Moreover, although the case where a metal was tabular was tried, various configurations, such as the shape of sponge, are also possible.
[0035] (Example 2) Although the conductive separator in which the gold plate layer was formed on the surface of th

metal was used in the example 1, this example shows the example in which the silver deposit was formed. In additio in this example, all of the plating approach, a cell configuration, and the characterization conditions of a cell presupposed that it is the same as that of an example 1.

[0036] A cell property the hydrogen gas humidified and warmed so that a fuel cell might be held at 85 degrees C and 83-degree C dew-point might come as well as an example 1 at one electrode side The air humidified and warmed wa supplied so that a 78-degree C dew-point might come at another electrode side, and the cell output when the first stag and operation time when performing a continuation generation-of-electrical-energy trial on condition that 80% of fue utilization rates, 40% of ratios of oxygen utilization, and current density 0.5 A/cm2 pass for 8000 hours was shown. The result was shown in Table 1.

[0037]

[Table 1]

電池	表面層	出力 (W)	
		初期	8000時間後
実施例1	金メッキ	1200	1000
実施例2	銀メッキ	1100	850
実施例3	金メッキ／金ペースト	1250	1080
実施例4	金ペースト	1080	950

[0038] (Example 3) Although the conductive separator in which the gold plate layer was formed on the surface of th metal was used in the example 1, this example shows the example which formed further the golden paste layer which has an organic binder component on it, and formed the conductive layer which has physical elasticity into the surface part of a conductive separator. In addition, in this example, all of the plating approach, a cell configuration, and the characterization conditions of a cell presupposed that it is the same as that of an example 1.

[0039] The formation approach of a golden paste layer of having an organic binder component distributed 50 % of th weight of gold grains of 20 micrometers of mean diameters to 10 % of the weight of bisphenol system epoxy monomers, and 10 % of the weight of the curing agent and the admixture of 50 % of the weight of ethyl Cellosolve which is a diluent, and after applying on the gold plate layer in which this was formed on the conductive separator, it performed them by leaving it at 120 degrees C for 5 hours.

[0040] A cell property the hydrogen gas humidified and warmed so that a fuel cell might be held at 85 degrees C and 83-degree C dew-point might come as well as an example 1 at one electrode side The air humidified and warmed wa supplied so that a 78-degree C dew-point might come at another electrode side, and the cell output when the first stag and operation time when performing a continuation generation-of-electrical-energy trial on condition that 80% of fue utilization rates, 40% of ratios of oxygen utilization, and current density 0.5 A/cm2 pass for 8000 hours was shown. The result was shown in Table 1.

[0041] (Example 4) In the above example, it gold-plated and the golden paste layer which has flexibility on it further was formed in the conductive separator made from SUS316. However, it is also possible to form directly on a conductive separator to adjust the component of a golden paste layer and made from SUS316. On the occasion of the quality governing of a golden paste layer at this time, if there are not much few golden components, SUS316 will be directly put to an external ambient atmosphere, and corrosion resistance falls as a conductive separator over a long period of time. On the other hand, if there are too many golden components, a binder component will fall conversely and the technical problem that flexibility is lost will happen. Moreover, naturally it is necessary to also optimize the particle size in the end of gold dust according to this. Electric resistance becomes small so that particle size is small, when not much small, the technical problem said that the treatment as a paste becomes difficult occurs.

[0042] After distributing 80 % of the weight of gold grains of 5 micrometers of mean diameters to 10 % of the weigh of bisphenol system epoxy monomers, and 5 % of the weight (carboxyl methyl cellulose) of CMC and the admixture 50 % of the weight of ethyl Cellosolve which is a diluent and applying this directly on a conductive separator by the above-mentioned production process in consideration of this point, the stability which excelled [degrees C / 120] in the thing performed by leaving it for 5 hours as well as the above was shown. [10 % of the weight of that curing age and] The configuration of other parts is the same as that of an example 1.

[0043] A cell property the hydrogen gas humidified and warmed so that a fuel cell might be held at 85 degrees C and 83-degree C dew-point might come as well as an example 1 at one electrode side The air humidified and warmed wa supplied so that a 78-degree C dew-point might come at another electrode side, and the cell output when the first stag and operation time when performing a continuation generation-of-electrical-energy trial on condition that 80% of fue utilization rates, 40% of ratios of oxygen utilization, and current density 0.5 A/cm2 pass for 8000 hours was shown. The result was shown in Table 1.

[0044]

[Effect of the Invention] Since according to this invention it can use without replacing the cutting method of construction of the conventional carbon plate as a separator plate and carrying out cutting of the metallic materials, such as stainless steel, large cost reduction can be planned at the time of mass production. Moreover, since a separato can be made much more thin, it contributes to miniaturization of a layer built cell.

[Translation done.]

CLAIMS

[Claim(s)]

[Claim 1] The electrode of the pair which sandwiches the solid-state polyelectrolyte film The means which carries ou supply discharge of the fuel gas at said electrode It is the solid-state polyelectrolyte mold fuel cell equipped with the above, and said conductive separator becomes with the metallic material in_which the inactive metal layer to an acid ambient atmosphere was formed on the front face, and said conductive separator forms the gas circulation slot which circulates said fuel gas, and it is characterize by to connect the means which carries out supply discharge of said gas circulation slot and said fuel gas with the ingredient which has the gas-seal nature to said fuel gas further.

[Claim 2] The inactive metal layer metal layer to the acid ambient atmosphere formed in the front face of a conductiv separator is a solid-state polyelectrolyte mold fuel cell according to claim 1 characterized by consisting of gold or silver.

[Claim 3] The inactive metal layer metal layer to the acid ambient atmosphere formed in the front face of a conductiv separator is a solid-state polyelectrolyte mold fuel cell according to claim 1 or 2 characterized by having physical flexibility.

[Claim 4] The solid-state polyelectrolyte mold fuel cell according to claim 1, 2, or 3 with which the gas circulation s formed in the conductive separator is characterized by being two or more parallel straight-line configurations mutual

[Claim 5] The solid-state polyelectrolyte mold fuel cell according to claim 1, 2, 3, or 4 with which the heights of the gas circulation slot formed in one field of a conductive separator are characterized by forming the concave of a gas circulation slot at the tooth back of said conductive separator.

[Claim 6] The manufacture approach of the solid-state polyelectrolyte mold fuel cell according to claim 2, 3, 4, or 5 characterized by having washing down stream processing, an activation process, nickel strike plating down stream processing, and gold plate or silver plating down stream processing.

[Translation done.]